

## **Title: NEUTRON IRRADIATION INDUCED METASTABLE STRUCTURES**

**Principal Investigator: Kurt Sickafus**

**KC0201020: FY01 budget: \$557 K**

**Project Summary:** The goal of this program is to understand the radiation damage response of ceramics exposed to neutrons or other energetic particles. Our studies of the damage response of ceramics address two objectives: (1) to predict microstructural evolution in ceramics exposed to radiation; and (2) to identify the physical aspects of ceramics that are effective in promoting radiation tolerance. Our ultimate goal is to design new radiation resistant ceramics. We conduct particle irradiation tests on ceramics to evaluate their irradiation damage response. We also perform computer simulations of defect energies and damage evolution in ceramics, to assist in our understanding of radiation damage phenomena in these materials. Our research is focused on highly radiation-resistant ceramics. We have determined that damage accumulation in oxides such as cubic-stabilized ZrO<sub>2</sub> and other fluorite-structured oxides, occurs at far lower rates than in most other ceramic oxides. We expect radiation tolerant ceramics to find application in existing fission reactors, in future fusion reactors or accelerator-based reactors, or as actinide-host ceramic fuel forms and waste forms.

### **Review Presentation Abstract:**

#### **RADIATION RESISTANCE IN A<sub>2</sub>O<sub>3</sub>-BO<sub>2</sub> OXIDES**

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Much work has been devoted in recent years to identifying ceramic materials that can withstand high doses of radiation without incurring excessive defect accumulation, or suffering undesirable transformations such as amorphization. In this presentation, we propose that a large range of A<sub>2</sub>O<sub>3</sub>-BO<sub>2</sub> oxide compositions, with structures related to the fluorite crystal structure, may exhibit exceptional resistance to radiation-induced amorphization. Results of heavy ion irradiations and calculations of defect energies for selected A<sub>2</sub>O<sub>3</sub>-BO<sub>2</sub> oxide compounds, are presented in support of this prediction.

### **Project Highlights:**

- Atomistic computer simulations along with ion beam irradiation experiments demonstrate that a group of compounds with chemical formulae given by A<sub>2</sub>B<sub>2</sub>O<sub>7</sub> and with crystal structures similar to that of the mineral fluorite are highly resistant to displacive radiation damage.

- Ion irradiation experiments reveal that  $\text{BO}_2$  compounds with the monoclinic baddeleyite structure (particularly  $\text{ZrO}_2$ ), transform to a structure similar to a cubic defective fluorite, and then persist without amorphization to very high ion dose.
- Ion irradiation experiments indicate that  $\text{A}_2\text{O}_3$  compounds with the cubic bixbyite structure (e.g.,  $\text{Dy}_2\text{O}_3$  and  $\text{Er}_2\text{O}_3$ ) transform to a structure similar to a cubic defective fluorite, and then persist without amorphization to very high ion dose.
- We have approximately 40 papers published between 1998 – 2001.

**Project Impact:** *Awards and Honors:* K. Sickafus: elected fellow of the American Ceramic Society (1998); elected into The Böhmische Physical Society (2000). *Conference Organization:* K. Sickafus: “Spinel Compounds: Structure Property Relations.” Annual Meeting of the American Ceramic Society, May, 1998, Cincinnati, OH (Journal of the American Ceramic Society (Vol. 82(12) December, 1999); K. Sickafus: “Structure-Property Relationships of Oxide Surfaces and Interfaces, MRS Fall Meeting, Dec., 2000, Boston, MA (MRS Proceedings, vol. 654, 2001); K. Sickafus: “Radiation-induced Effects by High Energy Particle/Solid Interaction,” 4<sup>th</sup> Pacific Rim International Conference on Advanced Materials and Processing, Dec., 2001, Honolulu, HI. *Degrees Awarded:* F. Li, New Mexico Institute of Mining and Technology, Socorro, NM, M.S. Thesis: “Radiation Damage on Rutile  $\text{TiO}_2$  Single Crystal,” May, 2000; L. Minervini, Imperial College, London, U. K., Ph.D. Thesis: “Atomistic Simulation of Defective Oxides, October, 2000; I. Afanasyev, Kharkiv National University, Kharkiv, Ukraine, Ph.D. Thesis: “Point Defects and Phase Transformations in Spinel and Zirconia Under Irradiation,” July, 2001.

**Project Interactions:** *External Collaborations:* R. W. Grimes, Imperial College, U. K.; HJ. Matzke, Transuranium Institute, Karlsruhe, GERMANY; Manabu Ishimaru, Osaka University; Osaka, JAPAN; V. T. Gritsyna, Kharkiv State University, Kharkiv, UKRAINE (In August, 2000, Gritsyna and Sickafus were awarded a collaborative research grant from the United States Civilian Research and Development Foundation (CRDF) entitled “Spatial Distribution and Interaction of Defects in Magnesium Aluminate Spinel, Based on Kinetic Studies of Irradiation Induced Processes Using Optical Methods.”). *Internal Collaborations:* LANL Accelerator Transmutation of Waste (ATW) Program with Ken McClellan and Ken Chidester. LANL Enhanced Surveillance Program on Pu Aging with Steve Valone, Marius Stan, Mike Nastasi, and Mike Baskes (MST-8).

**Project Time:** K. Sickafus, James Valdez, Ivan Afanasyev (MST-8)

Name:	Role:	% Time:	Status:
K. Sickafus	Staff Member, PI	80%	current
R. Grimes	Matthias Scholar	50%	Sept. 00 – Aug. 01
J. Valdez	Technician	70%	current
J. Williams	Undergraduate(U. Pittsburgh)	25%	May 01 – Aug. 01
I. Afanasyev	Graduate(Kharkiv National U.)	100%	Oct. 00 – Jan. 01

**Recent Publications:**

1. L. Minervini, R. W. Grimes and K. E. Sickafus, "Disorder in Pyrochlore Oxides," J. Am. Ceram. Soc. **83** (8) (2000) 1873-1878.
2. K. E. Sickafus, L. Minervini, R. W. Grimes, J. A. Valdez, M. Ishimaru, F. Li, K. J. McClellan and T. Hartmann, "Radiation tolerance of complex oxides," Science **289** (2000) 748-751.
3. M. Pirzada, R. W. Grimes, L. Minervini, J. F. Maguire and K. E. Sickafus, "Oxygen migration in  $A_2B_2O_7$  pyrochlores," Sol. St. Ionics **140** (2001) 201-208.
4. K. E. Sickafus, J. A. Valdez, J. R. Williams, R. W. Grimes and H. T. Hawkins, "Radiation Induced Amorphization Resistance in  $A_2O_3$ - $BO_2$  Oxides," Nucl. Instr. and Meth. in Phys. Res. B (2001) submitted for publication (Los Alamos Report # LA-UR-01-5577).